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(54) Title: SEPARATOR FOR LEAD-ACID STORAGE BATTERY (57) Abstract The electrical resistivity of a porous battery separator comprising: (a) a matrix consisting essentially of substantially water-insoluble thermoplastic organic polymer, at least 75 percent by weight of which is polyolefin, (b) finely divided amorphous silica particles distributed throughout the matrix, and (c) a network of interconnecting pores communicating substantially throughout the porous battery separator, wherein the porous battery separator has a silica to matrix polymer weight ratio in the range of from 2:1 to 3:1, may be reduced by treating the separators with corona discharge.		

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SEPARATOR FOR LEAD-ACID STORAGE BATTERY

Lead-acid storage batteries conventionally comprise acid electrolyte, at least one lead-containing anode and at least one lead-containing cathode at least partially immersed in said electrolyte and separated by a porous battery separator. The purpose of the battery separator is to prevent electrical shorts between anode and cathode while retaining acidic electrolyte in the pores for ionic conduction. Porous battery separators comprising amorphous silica and polyolefin are known and widely used in lead-acid storage batteries. See, for example, United States Patents No. 3,351,495; 3,798,294; 4,024,323; 4,210,709; 4,237,083; 4,331,622; 4,335,193; 4,648,417; 4,681,750; 4,734,229; and 5,605,569.

In general, battery separator manufacturers attempt to minimize electrical resistivity and maximize puncture strength for their separators. Most commercially available battery separators for lead-acid batteries have silica to matrix polymer weight ratios in the range of from 2:1 to 4:1 and porosities in the range of from 55 to 65 percent by volume. It is known that reducing the silica to matrix polymer weight ratio increases puncture strength, but such changes in formulation usually also result in higher electrical resistivity.

It has now been found that the electrical resistivity of porous battery separators having silica to matrix polymer weight ratios below 3:1 may be reduced by treating the separators with corona discharge. Surprisingly, no such reduction in electrical resistivity has been observed for porous battery separators having a silica to matrix polymer weight ratio of 3.5:1. This permits manufacturers to

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move toward lower silica to matrix polymer weight ratios in order to achieve greater puncture strengths without major increases in electrical resistivities.

Accordingly, in a porous battery separator
5 comprising: (a) a matrix consisting essentially of substantially water-insoluble thermoplastic organic matrix polymer, at least 75 percent by weight of which is polyolefin, (b) finely divided amorphous silica particles distributed throughout the matrix, and (c) a network of interconnecting
10 pores communicating substantially throughout the porous battery separator, one embodiment of the invention is the improvement wherein: (d) the porous battery separator has a silica to matrix polymer weight ratio in the range of from 2:1 to 3:1, and (e) the porous battery separator has been treated
15 with corona discharge.

Similarly, in a lead-acid storage battery comprising acid electrolyte, at least one lead-containing anode, and at least one lead-containing cathode, wherein the lead-containing anode and the lead-containing cathode are at least partially
20 immersed in said electrolyte and separated by a porous battery separator comprising: (a) a matrix consisting essentially of substantially water-insoluble thermoplastic organic matrix polymer, at least 75 percent by weight of which is polyolefin, (b) finely divided amorphous silica particles distributed
25 throughout the matrix, and (c) a network of interconnecting pores communicating substantially throughout the porous battery separator, another embodiment of the invention is the improvement wherein: (d) the porous battery separator has a silica to matrix polymer weight ratio in the range of from 2:1
30 to 3:1, and (e) the porous battery separator has been treated with corona discharge.

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Yet another embodiment of the invention is a method comprising treating with corona discharge a porous battery separator comprising: (a) a matrix consisting essentially of substantially water-insoluble thermoplastic organic matrix polymer, at least 75 percent by weight of which is polyolefin, (b) finely divided amorphous silica particles distributed throughout the matrix, and (c) a network of interconnecting pores communicating substantially throughout the porous battery separator, wherein the porous battery separator has a silica to matrix polymer weight ratio in the range of from 2:1 to 3:1.

Corona treatment of battery separators which do not contain silica, at least in the ratios claimed herein, and which are used in batteries other than lead-acid storage batteries, is known. However it appears that corona treatment of silica-containing battery separators, and the unexpected benefits to be attained through use of such corona-treated battery separators in lead-acid storage batteries have heretofore been unknown.

The batteries themselves (except for the corona discharge treated battery separators) are of conventional construction. Reference in this regard may be made to A. J. Salkind, J. J. Kelley and A. G. Cannone, Chapter 24, "Lead-Acid Batteries", in Handbook of Batteries, Second Edition, McGraw-Hill, Inc, New York, pages 24.1-24.89 (David Lindon, Editor-in Chief, 1995), Library of Congress Catalog No. TK2901.H36, the disclosure of which is, in its entirety, incorporated herein by reference.

Similarly, the battery separators before treatment with corona discharge are of conventional construction and may be produced by the methods disclosed in United States Patents No. 3,351,495; 3,798,294; 4,024,323; 4,210,709; 4,237,083;

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4,331,622; 4,335,193; 4,648,417; 4,681,750; 4,734,229; and 5,605,569, the disclosures of which are, in their entireties, incorporated herein by reference.

Each lead-containing electrode may independently
5 contain metallic lead, lead alloyed with other metals, or one or more lead compounds, or a combination of two or more thereof.

The acid electrolyte is conventional. It is aqueous and may contain one or more acids. Usually the electrolyte is
10 aqueous sulfuric acid having a specific gravity in the range of from 1.080 to 1.330. Often the specific gravity of the aqueous sulfuric acid is in the range of from 1.160 to 1.280 for batteries used for starting automobile engines. Minor amounts of various salts and additives may be present in the
15 electrolyte.

The polyolefin of the matrix polymer may be a homopolymer, a copolymer, a graft polymer, or a mixture of different polymers. Examples of suitable polyolefins include thermoplastic high density polyethylene, medium density
20 polyethylene, low density polyethylene, ultrahigh molecular weight polyethylene, polypropylene (atactic, isotactic, or syndiotactic as the case may be), copolymers of ethylene and propylene, copolymers of ethylene and butene. These listings are by no means exhaustive, but are intended for purposes of
25 illustration. The preferred polyolefins comprise substantially linear ultrahigh molecular weight polyolefin which is substantially linear ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 10 deciliters/gram, substantially linear ultrahigh molecular
30 weight polypropylene having an intrinsic viscosity of at least 6 deciliters/gram, or a mixture thereof. Substantially linear

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ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 18 deciliters/gram is especially preferred.

As used herein, intrinsic viscosity is determined by
5 extrapolating to zero concentration the reduced viscosities or the inherent viscosities of several dilute solutions of the UHMW polyolefin where the solvent is freshly distilled decahydronaphthalene to which 0.2 percent by weight, 3,5-di-~~tert~~- butyl-4-hydroxyhydrocinnamic acid,
10 neopentetetrayl ester [CAS Registry No. 6683-19-8] has been added. The reduced viscosities or the inherent viscosities of the UHMW polyolefin are ascertained from relative viscosities obtained at 135°C. using an Ubbelohde No. 1 viscometer in accordance with the general procedures of ASTM D 4020-81,
15 except that several dilute solutions of differing concentration are employed.

Nonolefinic thermoplastic polymer which is compatible with the polyolefin may optionally also be present in the matrix polymer. The amount of nonolefinic
20 thermoplastic polymer which may be present depends upon the nature of such polymer. In general, a greater amount of nonolefinic thermoplastic polymer may be used if its molecular structure contains little branching, few long sidechains, and few bulky side groups, than when there is a large amount of
25 branching, many long sidechains, or many bulky side groups.

Polyolefin constitutes at least 75 percent by weight of the matrix polymer. Usually polyolefin constitutes at least 90 percent by weight of the matrix polymer. Often polyolefin constitutes at least 95 percent by weight of the
30 matrix polymer. Preferably polyolefin constitutes substantially 100 percent by weight of the matrix polymer.

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The silica to matrix polymer weight ratio of the porous battery separator is in the range of from 2:1 to 3:1. Often the silica to matrix polymer weight ratio is in the range of from 2.2:1 to 3:1. Preferably the silica to matrix
5 polymer weight ratio is in the range of from 2.5:1 to 3:1.

Minor amounts, usually less than 15 percent by weight, of other materials used in processing such as lubricant, processing plasticizer (commonly referred to as "processing oil"), organic extraction liquid, water, and the
10 like, may optionally also be present in the battery separators. Other materials introduced for particular purposes may optionally be present in the battery separators in small amounts. Examples of such materials include antioxidants, ultraviolet light absorbers, reinforcing fibers
15 such as chopped glass fiber strand, dyes, fillers such as carbon black, and the like. Materials exemplified by the foregoing types may collectively be regarded as additives. The balance of the battery separators exclusive of filler is usually thermoplastic organic polymer.

20 Although the presence of processing plasticizer is optional, battery separators often contain from 10 to 15 percent processing plasticizer by weight.

The porosity of the battery separators of the invention can vary considerably, but usually the pores
25 constitute from 50 to 75 percent by volume of the porous battery separator. Preferably pores constitute from 55 to 65 percent by volume of the porous battery separator.

Apparatuses for producing corona discharge are numerous and well known, and their operation is well known.
30 The power levels, local intensities, and exposure times may vary widely. In general the exposure to corona discharge should be at least sufficient to achieve a meaningful reduction in electrical resistivity for the battery separator.

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As a battery separator containing amorphous silica and polyolefin in a silica to matrix polymer weight ratio in the range of from 2:1 to 3:1 is exposed to corona discharge, the electrical resistivity appears to approach a minimum value asymptotically. Continued exposure to corona discharge after the minimum value of electrical resistivity has been closely approached provides little added benefit. Further exposure seems not to be detrimental to the battery separator unless inordinately long exposures are employed. The maximum exposure to corona discharge is therefore not governed by scientific principles, but rather by non-technical considerations such as cost and convenience. Values of electrical resistivity as a function of corona discharge for any particular corona discharge treating apparatus and type of battery separator, may be quickly ascertained empirically using only a very few trials.

Other than in the operating examples, or where otherwise indicated, all numbers expressing quantities of ingredients or reaction conditions used herein are to be understood as modified in all instances by the term "about".

The invention is further described in conjunction with the following examples which are to be considered illustrative rather than limiting, and in which all parts are parts by weight and all percentages are percentages by weight unless otherwise specified.

EXAMPLES

Battery separators were extruded by mixing processing oil, precipitated silica, and ultrahigh molecular weight polyethylene (UHMWPE) in a batch mixer (see Table I). The precipitated silica served as the carrier for the oil phase such that a relatively dry, free flowing mixture was formed even as the oil content approached 60% by weight.

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The mixture was then fed into a twin screw extruder in which the barrel temperature was approximately 200°C. Additional oil was added in-line so that a homogeneous melt was formed. The melt was passed through a sheet die onto a calender roll in which the gap was used to control the extrudate thickness (~200 micrometers). The oil-filled separator was extracted with trichloroethylene to produce a battery separator containing approximately 12% residual oil by weight.

10

Table 1

Example No.	1	2 (Comparative)
Silica (1), g	454	454
PE/CB Concentrate (2), g	17.3	13.3
UHMWPE (3), g	168	128
Antioxidant (4), g	2.9	2.2
Lubricant (5), g	2.9	2.2
Processing Oil (6), g	788	797
SiO ₂ /MP (7)	2.7	3.5

20

Notes:

- (1) Hi-Sil[®] SBG Precipitated Silica, PPG Industries, Inc.
 (2) Polyblak[®] 1850V Polyethylene/Carbon Black Concentrate (Approximately 55% polyethylene, 45% carbon black, by weight), A. Schulmann Inc.
 (3) UHMWPE = Ultra High Molecular Weight Polyethylene, GUR 413, Hoechst-Celanese Corp.
 (4) Irganox[®] B-215, Ciba-Geigy Corp.
 (5) Petrac[®] CZ-81, Synpro Corp.
 (6) Shellflex[®] 3681, Shell Chemical Co.
 (7) SiO₂/MP = Silica/Matrix Polymer Weight Ratio.

After extraction with trichloroethylene and drying at room temperature, battery separators produced at silica to matrix polymer weight ratios of 2.7 and 3.5 were exposed to corona discharge using 3 or 5 passes of a wand hand-held about 2 millimeters above each side of the separator (Corona Discharge Unit Model No. BD-20; Electro-Technic Products Inc., Chicago, IL). The corona-treated battery separators

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were soaked in sulfuric acid (specific gravity = 1.280 at 27°C) and their electrical resistivity was measured in a Low Resistance Measuring System (Model #9100-2; Palico Instruments, Circle Pines, MN). The electrical resistivity values were measured after a 20 minute soak and a 24 hour soak in the sulfuric acid bath. The electrical resistivities of the corona-treated separators were compared to those of untreated control specimens (see Table 2).

10

Table 2

	<u>Example</u>	<u>SiO₂/MP</u>	<u>Corona Treatment</u>	<u>Electrical Resistivity,</u> <u>milliohm-meters</u>	
				<u>20 min soak</u>	<u>24 hr soak</u>
15	1	2.7	none	62.7	47.0
	1	2.7	3 passes	53.3	39.1
	1	2.7	5 passes	51.6	37.6
20	2	3.5	none	31.5	25.1
	2	3.5	3 passes	31.5	25.1
	2	3.5	5 passes	31.5	25.1

Although the present invention has been described with reference to specific details of certain embodiments thereof, it is not intended that such details should be regarded as limitations upon the scope of the invention except insofar as they are included in the accompanying claims.

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CLAIMS:

1. In a porous battery separator comprising:
 - (a) a matrix consisting essentially of substantially water-insoluble thermoplastic organic matrix polymer, at least 75 percent by weight of which is polyolefin,
 - (b) finely divided amorphous silica particles distributed throughout the matrix, and
 - (c) a network of interconnecting pores communicating substantially throughout the porous battery separator,the improvement wherein:
 - (d) the porous battery separator has a silica to matrix polymer weight ratio in the range of from 2:1 to 3:1, and
 - (e) the porous battery separator has been treated with corona discharge.
2. The porous battery separator of claim 1 wherein the polyolefin constitutes at least 90 percent by weight of the matrix polymer.
3. The porous battery separator of claim 1 wherein the polyolefin constitutes substantially 100 percent by weight of the matrix polymer.
4. The porous battery separator of claim 1 wherein the polyolefin comprises substantially linear ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 10 deciliters/gram, substantially linear ultrahigh molecular weight polypropylene having an intrinsic viscosity of at least 6 deciliters/gram, or a mixture thereof.

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5. The porous battery separator of claim 1 wherein the polyolefin comprises substantially linear ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 18 deciliters/gram.

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6. The porous battery separator of claim 1 wherein the silica to matrix polymer weight ratio is in the range of from 2.2:1 to 3:1.

10

7. The porous battery separator of claim 1 wherein the silica to matrix polymer weight ratio is in the range of from 2.5:1 to 3:1.

8. The porous battery separator of claim 1 which contains from 10 to 15 percent processing plasticizer by weight.

9. The porous battery separator of claim 1 wherein pores constitute from 50 to 75 percent by volume of the battery separator.

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10. The porous battery separator of claim 1 wherein pores constitute from 55 to 65 percent by volume of the battery separator.

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11. In a lead-acid storage battery comprising acid electrolyte, at least one lead-containing anode, and at least one lead-containing cathode, wherein the lead-containing anode and the lead-containing cathode are at least partially immersed in said electrolyte and separated by a porous battery separator comprising:

- (a) a matrix consisting essentially of substantially water-insoluble thermoplastic organic matrix polymer, at least 75 percent by weight of which is polyolefin,
- (b) finely divided amorphous silica particles distributed throughout the matrix, and
- (c) a network of interconnecting pores communicating substantially throughout the porous battery separator,

the improvement wherein:

- (d) the porous battery separator has a silica to matrix polymer weight ratio in the range of from 2:1 to 3:1, and
- (e) the porous battery separator has been treated with corona discharge.

12. The lead-acid storage battery of claim 11 wherein the polyolefin constitutes at least 90 percent by weight of the matrix polymer of the porous battery separator.

13. The lead-acid storage battery of claim 11 wherein the polyolefin constitutes substantially 100 percent by weight of the matrix polymer of the porous battery separator.

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14. The lead-acid storage battery of claim 11 wherein the polyolefin comprises substantially linear ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 10 deciliters/gram, substantially linear
5 ultrahigh molecular weight polypropylene having an intrinsic viscosity of at least 6 deciliters/gram, or a mixture thereof.

15. The lead-acid storage battery of claim 11 wherein the polyolefin comprises substantially linear
10 ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 18 deciliters/gram.

16. The lead-acid storage battery of claim 11 wherein the silica to matrix polymer weight ratio of the
15 porous battery separator is in the range of from 2.2:1 to 3:1.

17. The lead-acid storage battery of claim 11 wherein the silica to matrix polymer weight ratio of the porous battery separator is in the range of from 2.5:1 to 3:1.
20

18. The lead-acid storage battery of claim 11 wherein the porous battery separator contains from 10 to 15 percent processing plasticizer by weight.

25 19. The lead-acid storage battery of claim 11 wherein pores constitute from 50 to 75 percent by volume of the battery separator.

20. The lead-acid storage battery of claim 11
30 wherein pores constitute from 55 to 65 percent by volume of the battery separator.

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21. A method comprising treating with corona discharge a porous battery separator comprising:

- (a) a matrix consisting essentially of substantially water-insoluble thermoplastic organic matrix polymer, at least 75 percent by weight of which is polyolefin,
- (b) finely divided amorphous silica particles distributed throughout the matrix, and
- (c) a network of interconnecting pores communicating substantially throughout the porous battery separator,

wherein the porous battery separator has a silica to matrix polymer weight ratio in the range of from 2:1 to 3:1.

22. The method of claim 21 wherein the polyolefin constitutes at least 90 percent by weight of the matrix polymer of the porous battery separator.

23. The method of claim 21 wherein the polyolefin constitutes substantially 100 percent by weight of the matrix polymer of the porous battery separator.

24. The method of claim 21 wherein the polyolefin comprises substantially linear ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 10 deciliters/gram, substantially linear ultrahigh molecular weight polypropylene having an intrinsic viscosity of at least 6 deciliters/gram, or a mixture thereof.

25. The method of claim 21 wherein the polyolefin comprises substantially linear ultrahigh molecular weight polyethylene having an intrinsic viscosity of at least 18 deciliters/gram.

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26. The method of claim 21 wherein the silica to matrix polymer weight ratio of the porous battery separator is in the range of from 2.2:1 to 3:1.

5

27. The method of claim 21 wherein the silica to matrix polymer weight ratio of the porous battery separator is in the range of from 2.5:1 to 3:1.

10

28. The method of claim 21 wherein the porous battery separator contains from 10 to 15 percent processing plasticizer by weight.

29. The method of claim 21 wherein pores
15 constitute from 50 to 75 percent by volume of the battery separator.

30. The method of claim 21 wherein pores
constitute from 55 to 65 percent by volume of the battery
20 separator.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 H01M2/16 H01M2/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 095, no. 009, 31 October 1995 & JP 07 142047 A (MITSUBISHI PAPER MILLS LTD), 2 June 1995 see abstract ---	1-3, 21-23
A	PATENT ABSTRACTS OF JAPAN vol. 095, no. 008, 29 September 1995 & JP 07 134979 A (MATSUSHITA ELECTRIC IND CO LTD), 23 May 1995 see abstract ---	1
A	PATENT ABSTRACTS OF JAPAN vol. 017, no. 261 (E-1369), 21 May 1993 & JP 05 006760 A (NIPPON MUKI CO LTD), 14 January 1993 see abstract ---	1
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>US 4 335 193 A (DOI YOSHINAO ET AL) 15 June 1982 cited in the application see column 3, line 36 - line 38; claim 1 -----</p>	11

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 98/07782

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